

**Title: Controls on millennial-scale atmospheric CO<sub>2</sub> variability during the last glacial period**

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**Key points**

- A new ice core record of the stable isotopes of atmospheric CO<sub>2</sub> suggests organic carbon sources controlled CO<sub>2</sub> levels during the last glacial period
- The millennial-scale CO<sub>2</sub> variability is tentatively linked to variations in Southern Ocean carbon sources
- Centennial-scale CO<sub>2</sub> variability during the last glacial period is associated with similarly abrupt changes during the deglaciation

## Abstract

Changes in atmospheric CO<sub>2</sub> on millennial-to-centennial timescales are key components of past climate variability during the last glacial and deglacial periods (70-10ka) yet the sources and mechanisms responsible for the CO<sub>2</sub> fluctuations remain largely obscure. Here we report the <sup>13</sup>C/<sup>12</sup>C ratio of atmospheric CO<sub>2</sub> during a key interval of the last glacial period at sub-millennial resolution, with coeval histories of atmospheric CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O concentrations. The carbon isotope data suggest that the millennial-scale CO<sub>2</sub> variability in MIS3 is driven largely by changes in the organic carbon cycle, most likely by sequestration of respired carbon in the deep ocean. Centennial-scale CO<sub>2</sub> variations, distinguished by carbon isotope signatures, are associated with both abrupt hydrological change in the tropics (e.g. Heinrich Events) and rapid increases in northern hemisphere temperature (DO events). These events can be linked to modes of variability during the last deglaciation, thus suggesting that drivers of millennial and centennial CO<sub>2</sub> variability during both periods are intimately linked to abrupt climate variability.

## Plain Language Summary

Ice cores provide unique records of variations in atmospheric CO<sub>2</sub> prior to the instrumental era. While it is clear that changes in atmospheric CO<sub>2</sub> played a significant role in driving past climate change, it is unclear what in turn drove changes in atmospheric CO<sub>2</sub>. Here we investigate enigmatic changes in atmospheric CO<sub>2</sub> levels during an interval of the last glacial period (~50,000 to 35,000 years ago) that are associated with abrupt changes in polar climate. To determine the sources and sinks for atmospheric CO<sub>2</sub> we measured the stable isotopes of carbon in CO<sub>2</sub> and found that the primary source of carbon to the atmosphere was an organic carbon reservoir. Most likely, this carbon was sourced from a deep ocean reservoir that waxed and waned following changes in either the productivity of the surface ocean or stratification of the deep ocean. We also found that atmospheric CO<sub>2</sub> can change on the centennial time-scale during abrupt climate transitions in the Northern Hemisphere. This observation adds to a growing body of evidence that abrupt changes in atmospheric CO<sub>2</sub> are an important component of past carbon cycle variability.

## 73 Introduction

74 In order to predict the climate impacts of anthropogenic CO<sub>2</sub> emissions over the coming millennia  
75 (Clark et al., 2016) we must understand how the climate system and carbon cycle have interacted over  
76 these same timescales in the past. The CO<sub>2</sub> rise during the last deglaciation, arguably the most well-  
77 studied example of past carbon cycle variability, is likely a combination of millennial-scale climate  
78 change and glacial-interglacial shifts in temperature and ice volume, which are all amplified through a  
79 system of climate-carbon cycle feedbacks. To disentangle the millennial-scale component we  
80 investigate carbon cycle variability of the last glacial period when climate variations were largely  
81 unaffected by changes in northern hemisphere insolation and ice volume that characterize glacial  
82 terminations (Figure S1).

83

84 During Marine Isotope Stage 3 (MIS3) atmospheric CO<sub>2</sub> varied between about 195 and 225 ppm with a  
85 roughly triangular waveform (Indermühle, 2000) (Figure 1 E). This pattern mimics Antarctic  
86 temperature, rising during the strongest Antarctic warmings and falling during the coolings (Figure 1  
87 B). From the perspective of Greenland climate and the well-known Dansgaard-Oeschger events (DO),  
88 CO<sub>2</sub> increases during cold phases (stadials)(Ahn & Brook, 2008) with the most prominent increases  
89 corresponding to only those stadials associated with an enhanced flux of debris-laden ice to the North  
90 Atlantic (known as a Heinrich Events; these specific cold intervals are often referred to as “Heinrich  
91 stadials”). CO<sub>2</sub> decreases after a rapid switch to warmer conditions over Greenland (interstadials) and  
92 slower cooling in Antarctica (Bereiter et al., 2012). This strong relationship between Antarctic  
93 temperature and CO<sub>2</sub> has focused attention on the Southern Ocean (SO) as the major conduit for the  
94 transfer of carbon between the atmosphere and the ocean on millennial and glacial-interglacial  
95 timescales (Sigman et al., 2010), with possible triggers in the North Atlantic.

96

97 The ratios of carbon isotopes differ among key carbon reservoirs and thus some of the sources and  
98 sinks driving past atmospheric CO<sub>2</sub> can be constrained using the isotopic composition of atmospheric  
99 CO<sub>2</sub> (Bauska et al., 2016; Schmitt et al., 2012; Köhler et al., 2006; Eggleston et al., 2016). Four major  
100 processes fractionate the carbon isotopes ( $\epsilon_{A-B} \approx \delta^{13}C_A - \delta^{13}C_B$ ): photosynthesis that makes CO<sub>2</sub> into  
101 organic carbon on land ( $\epsilon_{\text{land-atmosphere}} \approx -18\text{‰}$ ), photosynthesis in the surface ocean that forms  
102 particulate organic carbon from dissolved inorganic carbon ( $\epsilon_{\text{DIC-POC}} \approx -22\text{‰}$ ), air-sea gas exchange  
103 ( $\epsilon_{\text{DIC-atmosphere}} \approx +8\text{‰}$ ; decreasing by 0.1‰ for every 1°C increase in ocean temperature), and a  
104 negligible fractionation during the formation of CaCO<sub>3</sub> in surface ocean, ( $\epsilon_{\text{DIC-CaCO}_3} \approx 0\text{‰}$ ). By  
105 measuring the time varying changes in both atmospheric CO<sub>2</sub> concentrations and the  $\delta^{13}C$  isotopes, we  
106 can constrain the sources of the CO<sub>2</sub> changes. Rising atmospheric CO<sub>2</sub> and decreasing  $\delta^{13}C$ -CO<sub>2</sub> is  
107 consistent with organic carbon sources to the atmosphere; rising atmospheric CO<sub>2</sub> and increasing  
108  $\delta^{13}C$ -CO<sub>2</sub> is consistent with a warming ocean; and rising CO<sub>2</sub> with no changes in  $\delta^{13}C$ -CO<sub>2</sub> could indicate  
109 a balanced contribution of rising ocean temperature and organic carbon sources or the influence of

110  $\text{CaCO}_3$  or volcanic sources. Air-sea gas exchange in the high-latitude ocean may affect atmospheric  
111  $\delta^{13}\text{C}\text{-CO}_2$ , with rising atmospheric  $\text{CO}_2$  and large decreases in  $\delta^{13}\text{C}\text{-CO}_2$  predicted by box model  
112 experiments that include enhanced air-sea gas exchange in the SO (Bauska et al., 2016; Köhler et al.,  
113 2006). Finally, changes in the abundance of C3 and C4 plants on land (Ehleringer et al., 1997) or  
114 ecological shifts in marine biosphere (Broecker & McGee, 2013) would change the overall  
115 photosynthetic fractionation and thus the details, but not the fundamentals, of using  $\delta^{13}\text{C}\text{-CO}_2$  to  
116 fingerprint carbon sources.

117

118 Other atmospheric gases provide additional constraints on carbon cycle variability, particularly  
119 related to the response of the terrestrial biosphere to abrupt climate change. Most evidence suggests  
120 that past variations in methane are dominated by climate variability over tropical and boreal wetlands  
121 (Brook et al., 2000; Rhodes et al., 2015).  $\text{N}_2\text{O}$  is produced in oxygen-poor ocean waters and terrestrial  
122 soils. Rising  $\text{N}_2\text{O}$  most likely reflects either decreasing oxygen levels in the intermediate-depth ocean,  
123 warmer terrestrial soil temperatures or a combination of the two (Schilt et al., 2014). The  $\delta^{18}\text{O}\text{-O}_2$   
124 largely follows the changes in  $\delta^{18}\text{O}$  of seawater and orbitally-driven changes in the Dole effect, but  
125 rapid increases in  $\delta^{18}\text{O}\text{-O}_2$  have been argued to reflect southward shifts in low-latitude precipitation  
126 (Seltzer et al., 2017; Severinghaus et al., 2009) (see SI for definition of  $\Delta\epsilon_{\text{LAND}}$ ).

127

## 128 **Methods and Materials**

129 We present new atmospheric histories of  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{CH}_4$  and  $\delta^{13}\text{C}\text{-CO}_2$  spanning 47-35 ka from the  
130 Taylor Glacier, Antarctica blue ice area (Figure 1 D-G). The chronology (Baggenstos et al., 2017) is  
131 constructed by synchronizing the Taylor Glacier  $\text{CH}_4$  and  $\delta^{18}\text{O}\text{-O}_2$  records to the WDC timescale  
132 (Buizert et al., 2015) and thus linking to the radiometrically dated Hulu Cave record (Cheng et al.,  
133 2016) (Figure 1 C). The concentrations of  $\text{CO}_2$  and  $\text{N}_2\text{O}$  and the  $\delta^{13}\text{C}\text{-CO}_2$  were measured on the same  
134 sample using the Oregon State University ice grater system (Bauska et al., 2014). The long-term  
135 reproducibility for  $\text{CO}_2$ ,  $\text{N}_2\text{O}$  and  $\delta^{13}\text{C}\text{-CO}_2$  are  $\pm 1\text{ppm}$ ,  $\pm 5\text{ppb}$ ,  $+0.02\text{‰}$ , respectively (Bauska et al., 2015;  
136 Bauska et al., 2016). The combination of increased resolution and precision equates to a significant  
137 improvement over previous reconstructions which focused on longer-term changes (Eggleson et al.,  
138 2016) (Figure S1). Additionally, previous reconstructions in this time interval were limited by offsets  
139 between cores (see *Eggleson et al., 2016* for detailed discussion and Figure S2). Using the interval  
140 from 47 to 43ka as a baseline for comparison, the Talos Dome  $\delta^{13}\text{C}\text{-CO}_2$  record has significantly lower  
141 and more variable values (mean $\pm$ s.d. =  $-6.72\pm 0.23\text{‰}$ ;  $n = 12$ ) than data from the EDML ( $-6.50\pm 0.12\text{‰}$ ;  
142  $n = 8$ ) and EDC ( $-6.59\pm 0.16\text{‰}$ ;  $n = 5$ ) ice cores. In the same interval, the Taylor Glacier averages -  
143  $6.55\pm 0.07\text{‰}$  ( $n = 14$ ) in broad agreement with EDML and EDC. Although we can confidently interpret  
144 relative changes in our new record because of the improvement in precision and resolution,  
145 addressing the absolute accuracy of the  $\delta^{13}\text{C}\text{-CO}_2$  values requires additional inter-laboratory and inter-  
146 core comparison.

147

148 **Results: Ice Core Constraints on Greenhouse Gas Variability**

149 The most salient mode of variability in atmospheric CO<sub>2</sub>, the millennial-scale rising and falling with  
150 Antarctic temperature, is accompanied by an inversely correlated change in δ<sup>13</sup>C-CO<sub>2</sub> (Figure 1 E-F).  
151 Atmospheric CO<sub>2</sub> ranges from about 195 to 215 ppm with the corresponding variability in δ<sup>13</sup>C-CO<sub>2</sub>  
152 spanning -6.45 to -6.65‰ (a -0.1‰ change for every +10 ppm). At finer scales we observe several  
153 other modes of variability. The CO<sub>2</sub> rise during Heinrich Stadial 4 (40.2-38.4 ka; “HS4”) starts off  
154 slowly, rising 3-4 ppm while δ<sup>13</sup>C-CO<sub>2</sub> decreases by ~0.05‰. As noted previously in the Siple Dome ice  
155 core (Ahn & Brook, 2014), the rise then accelerates in a sharp jump of about 8 ppm. The rapid phase of  
156 the rise is coincident with the mid-stadial rise in CH<sub>4</sub> noted in the WAIS Divide Core (Rhodes et al.,  
157 2015) (Figure 1 D, red vertical line) and the increase in Δε<sub>LAND</sub> first observed in the Siple Dome Core  
158 (Severinghaus et al., 2009) and confirmed in the WDC record (Seltzer et al., 2017) (Figure 1 H). No  
159 change in N<sub>2</sub>O is resolved in the Taylor Glacier dataset, consistent with the Talos Dome record (Schilt  
160 et al., 2010) (Figure 1 G). The resolution of the carbon isotope measurements prevents a clear  
161 fingerprinting of the source but a replicated sample clearly falls to more negative values off the more  
162 gradual trend by about 0.1‰ (Figure 1 F). In the later part of the HS4, CO<sub>2</sub> continues to rise slowly by  
163 3 ppm and δ<sup>13</sup>C-CO<sub>2</sub> decreases by another 0.1‰.

164

165 The onsets of DO interstadials are accompanied by small rises in CO<sub>2</sub> (Figure 1 E, dashed gray lines).  
166 This variability has been noted in other cores and described as either a lagged response to Antarctic  
167 temperature (Bereiter et al., 2012) or, in the case of the weaker DO events, a lagged response to  
168 Greenland stadial conditions that are too short to impart a significant change in CO<sub>2</sub> (Ahn & Brook,  
169 2014). In our record we note that atmospheric CO<sub>2</sub> appears to increase along with CH<sub>4</sub>, and N<sub>2</sub>O with  
170 no discernable lead or lag. This is most prominent at DO8 when CH<sub>4</sub> rises by about 120 ppb, N<sub>2</sub>O rises  
171 by 35 ppb and CO<sub>2</sub> rises by 6 ppm (see also Figure 4 n-p). The other DO events (7,9,10) are near the  
172 detection limit of our record and difficult to quantify. Across these events δ<sup>13</sup>C-CO<sub>2</sub> either increases  
173 slightly (~0.08‰ at DO8) or shows no substantial change.

174

175 **Discussion: Millennial-scale carbon cycle variability**

176 The overall negative correlation between CO<sub>2</sub> and δ<sup>13</sup>C-CO<sub>2</sub> rules out changes in ocean temperature,  
177 the CaCO<sub>3</sub> cycle, or volcanic input having a dominant role in driving millennial-scale CO<sub>2</sub> (Figure 2).  
178 Instead, the data are consistent with changes in terrestrial carbon storage or the strength of the  
179 ocean’s biological pump. If terrestrial sources were dominant, whole ocean δ<sup>13</sup>C would follow  
180 atmospheric δ<sup>13</sup>C-CO<sub>2</sub>. If oceanic sources from changes in the strength of the biological pump or shifts  
181 in ocean ventilation were dominant, the surface-to-deep gradient in δ<sup>13</sup>C in inorganic carbon in the  
182 ocean would decrease along with atmospheric δ<sup>13</sup>C-CO<sub>2</sub>.

183

184 Millennial-scale decreases in the vertical gradient of  $\delta^{13}\text{C}$  in the SO have been tentatively correlated to  
185 maxima in atmospheric  $\text{CO}_2$  (Charles et al., 2010; Ziegler et al., 2013), and therefore in light of our new  
186 data, minima in  $\delta^{13}\text{C}\text{-CO}_2$  (Figure 3a-b). If this coupling of the oceanic gradient of  $\delta^{13}\text{C}$  and atmospheric  
187  $\delta^{13}\text{C}\text{-CO}_2$  could be demonstrated to be precisely in-phase it would provide clear evidence of an oceanic  
188 source controlling atmospheric  $\text{CO}_2$ . Taking the current data and chronology at face-value, the  
189 decreases in the oceanic  $\delta^{13}\text{C}$  gradient are broadly associated with minima in atmospheric  $\delta^{13}\text{C}\text{-CO}_2$ ,  
190 yet the coupling is clearly not one-to-one, possibly due to chronological errors in the marine sediment  
191 record. To explore this hypothesis further, we combined existing benthic carbon isotope records from  
192 deep SO ( $\sim 42^\circ\text{S}$ ,  $10^\circ\text{E}$ , 4600 m) (Charles et al., 1996; Ninnemann et al., 1999; Hodell et al., 2001, 2003).  
193 These records use previously established age models tied to the Greenland ice core records using  
194 carbonate preservation and confirmed by  $^{14}\text{C}$  data during the deglaciation and the Laschamp  
195 paleomagnetic event during MIS3 ( $\sim 42\text{ka}$ ) (Barker & Diz, 2014). We include a minor increase in the  
196 absolute age of 0.63% to account for the possible under-counting of annual layers in the Greenland Ice  
197 Core chronologies relative to the WAIS Divide timescale (Buizert et al., 2015).

198  
199 We note that the atmosphere and deep ocean  $\delta^{13}\text{C}$  are anti-correlated during MIS3 (Figure 3c). This  
200 supports the hypothesis that the waxing and waning of respired organic carbon source in the deep SO  
201 controlled atmospheric  $\text{CO}_2$  and significantly limits the possibility of contributions from terrestrial  
202 sources. However, this relationship alone cannot delineate oceanic sources between changes in export  
203 productivity and changes in stratification. Evidence to support changes in export productivity comes  
204 from the correlation of the ice core data and foraminifera-bound  $\delta^{15}\text{N}$  which indicates lower nitrate  
205 utilization during periods of high atmospheric  $\text{CO}_2$  (Martínez-García et al., 2014) (Figure 3d). Iron  
206 deposition rates and export production in the Subantarctic are also closely coupled, suggesting that  
207 the extent of iron limitation may have played a role in this enhanced nutrient utilization (Jaccard et al.,  
208 2016). Evidence in support of ventilation changes stems from radiocarbon constraints in the South  
209 Atlantic that are closely coupled with deep ocean oxygen levels, suggesting that both export  
210 productivity and ocean circulation were working in concert over this period (Gottschalk et al., 2016).  
211 A quantitative description of this coupling requires study with isotope enabled earth system models;  
212 however, additional insight can be gained by comparing to periods when these processes may have  
213 become uncoupled.

214  
215 Our new high-resolution MIS3 data now provide a similar sequence of abrupt climate change events to  
216 contrast with the last deglaciation. First, we compare the MIS3 data to the last deglaciation utilizing a  
217 cross plot of  $\text{CO}_2$  and  $\delta^{13}\text{C}\text{-CO}_2$  (Figure 2). We see that the MIS3 data fall along the same trend observed  
218 during HS1 of the last deglaciation ( $\sim 18\text{-}15.0\text{ka}$ ) yet only span about 50% of range (note that the low  
219  $\delta^{13}\text{C}\text{-CO}_2$  values that fall off this trend are due to centennial-scale variability at  $\sim 16.3\text{ ka}$ ). This suggests  
220 that millennial-scale  $\text{CO}_2$  variations in MIS3 can be linked mechanistically to the more pronounced

221 variability during the deglaciation. As discussed in previous work, this co-variability of CO<sub>2</sub> and δ<sup>13</sup>C-  
 222 CO<sub>2</sub> is consistent with changes in SO ventilation (Köhler et al., 2006; Menviel et al., 2015; Schmitt et al.,  
 223 2012; Tschumi et al., 2011) changes in export production (particularly the SO)(Bauska et al., 2016;  
 224 Menviel et al., 2012) or a weakening of the ocean's biological pump by a reduction in North Atlantic  
 225 Deep-water (NADW) formation (Schmittner & Lund, 2015).  
 226  
 227 Second, we compare the two intervals in time with the GHG records plotted over 8ka intervals (Figure  
 228 4a-c). Similar patterns of millennial-scale variability in CH<sub>4</sub> and N<sub>2</sub>O are observed but the changes in  
 229 CO<sub>2</sub> appear fundamentally different. First, the CO<sub>2</sub> rise during HS1 is about 20 ppm greater than the  
 230 rise in HS4 (or ~2x) and, second, CO<sub>2</sub> remains elevated after the switch to interstadial conditions  
 231 during the Bølling-Allerød (BA) rather than decreasing as observed after the onset of DO8 (Figure 3 C).  
 232 The δ<sup>13</sup>C-CO<sub>2</sub> follows a similar pattern. In HS1 δ<sup>13</sup>C-CO<sub>2</sub> decreases by 0.2‰ more than HS4 and  
 233 stabilizes during the BA as opposed to abrupt increases observed during DO8 (Figure 4 D). What was  
 234 different about the deglaciation that allowed more respired carbon into the atmosphere during HS1  
 235 than HS4 and prevented, or compensated for, a potential uptake of carbon during the BA?  
 236  
 237 During the Heinrich stadials, NADW weakens (Henry et al., 2016) and Subantarctic productivity  
 238 decreases (Anderson et al., 2014; Gottschalk et al., 2016) tracking dust delivery to Antarctica (Fischer  
 239 et al., 2007) (Figure 4 e, h-i). Ventilation of the SO (as inferred from radiocarbon) improves  
 240 (Gottschalk et al., 2016; Skinner et al., 2010) and SO upwelling (as inferred from Antarctic  
 241 productivity) increases (Anderson et al., 2009), although these changes could be more pronounced in  
 242 the later part of the Heinrich stadials (Figure 4 f-g). During MIS3 all of these changes are largely  
 243 symmetric around, and consistent with, the minimum in δ<sup>13</sup>C-CO<sub>2</sub> and thus plausible drivers the  
 244 change in CO<sub>2</sub>.  
 245  
 246 During the deglaciation some variables trend back towards LGM values after HS1 (NADW and SO  
 247 upwelling), while others, show near permanent shifts to interglacial levels during HS1 (Subantarctic  
 248 productivity, SO ventilation). This decoupling allows us to partially disentangle which processes  
 249 control atmospheric CO<sub>2</sub>. Based on relationship between the proxies and atmospheric data in MIS3,  
 250 we would expect that if changes in NADW and/or SO upwelling were to control atmospheric CO<sub>2</sub>, we  
 251 would observe a large CO<sub>2</sub> decrease and δ<sup>13</sup>C-CO<sub>2</sub> increase in the BA. This is clearly not the case and  
 252 requires either a muted response to these forcings or another source of carbon in the BA that  
 253 compensated for the apparent carbon sink. Conversely, if changes in Subantarctic productivity and SO  
 254 ventilation dominated the atmospheric CO<sub>2</sub> budget, we could expect CO<sub>2</sub> to remain elevated during the  
 255 BA and δ<sup>13</sup>C-CO<sub>2</sub> to plateau at lower values, a scenario that is in much better agreement with the data.  
 256 Thus Subantarctic productivity and SO ventilation appear to have a more consistent link with

257 atmospheric CO<sub>2</sub> in both MIS3 and the Last Deglaciation and are strong candidates for contributing  
258 significantly to glacial-interglacial CO<sub>2</sub> change.

259

## 260 **Discussion: Centennial-scale carbon cycle variability**

261 On the centennial-scale our new observations in MIS3 can be combined with recently identified  
262 variability in Last Deglaciation to suggest a ubiquitous and consistent coupling of the carbon cycle with  
263 abrupt climate change events. In Figure 4j-q we plot variability in greenhouse gases for two categories  
264 of centennial-scale events: the onset of interstadials (D08, the Bølling-Allerød and the pre-Boreal) and  
265 mid-Heinrich Stadials events (H4 and H1). The changes are plotted as anomalies and the timing is set  
266 relative to the mid-point of the rise in CH<sub>4</sub>. Note that the Taylor Glacier chronology is synchronized  
267 with the WAIS Divide record via CH<sub>4</sub> and thus the phasing of the two ice cores cannot be interpreted.

268

269 The rapid 8 ppm CO<sub>2</sub> rise during HS4 at 39.5 ka likely shares a common origin with a similar event  
270 during the deglaciation with HS1 at 16.3 ka (Marcott et al., 2014) (Figure 4l). Both events are  
271 associated with carbon isotope minima (Figure 4m). It has been suggested that these mid-stadial  
272 events can be tied to the timing of the Heinrich Events and may represent a rapid release of terrestrial  
273 carbon to the atmosphere driven by a cooling and drying of the Northern Hemisphere (Bauska et al.,  
274 2016). The event in HS4 is consistent with a terrestrial origin as it coincides with an increase in CH<sub>4</sub>  
275 (Rhodes et al., 2015) (Figure 4j), which may indicate enhanced precipitation of the Southern  
276 Hemisphere tropics, and an increase in  $\Delta\epsilon_{\text{LAND}}$ , which suggests decreased precipitation in the Northern  
277 Hemisphere (Seltzer et al., 2017; Severinghaus et al., 2009) (Figure 1h). Constant N<sub>2</sub>O indicates either  
278 that changes in terrestrial soil temperatures may have been small on the global scale, thus suggesting  
279 that precipitation was the dominant driver of the terrestrial carbon loss, or that oxygen-minimum  
280 zones in intermediate ocean were relatively stable, possibly indicating the absence of a change ocean  
281 circulation across this event (Figure 4k).

282

283 The 6 ppm CO<sub>2</sub> rise at the onset of D08 shares common features with the onset of the BA and  
284 Preboreal (~11.5ka) during the deglaciation (Figure 4p). All three events exhibit simultaneous  
285 increases in CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O that coincide with abrupt Northern Hemisphere warmings, continued  
286 warming or at least stable temperatures in Antarctica (WAIS Divide Project Members, 2015), and  
287 greater NADW formation (Henry et al., 2016; McManus et al., 2004) (Figure 3, vertical yellow dashed  
288 lines). The  $\delta^{13}\text{C}$ -CO<sub>2</sub> across all events is variable but shows no secular trend (Preboreal) or slight  
289 increase of ~0.08‰ (B/A and D08) (Figure 4q). At face value, this pattern of increasing CO<sub>2</sub> and  
290 increasing  $\delta^{13}\text{C}$ -CO<sub>2</sub> indicates that rising ocean temperature contributed to the CO<sub>2</sub> rise with additional  
291 (but limited) changes in the net flux of organic carbon. This simplest of scenarios is somewhat  
292 surprising given that changes in the ocean's biological pump may accompany the large and abrupt  
293 reorganization of ocean circulation and changes in terrestrial carbon reservoirs are clearly indicated by



the large increases in CH<sub>4</sub> and N<sub>2</sub>O (Figure 5e). More plausibly, yet more challenging to accurately model, are a roughly synchronous changes in several sources. Recently, the LOVECLIM model, which predicts a small positive relationship between CO<sub>2</sub> and δ<sup>13</sup>C-CO<sub>2</sub> during reduced NADW (Menviel et al., 2015), also predicts increases in CO<sub>2</sub> of 10 to 15 ppm upon the resumption of NADW (with the effect of solubility contributing about 50% to CO<sub>2</sub> variability) (Menviel et al., 2015). Moreover, a precisely dated coral record shows that these events during the last deglaciation are associated with brief intervals of enhanced overturning in the Atlantic (Chen et al., 2015). This integrated response to the onset of an interstadial is consistent with the CO<sub>2</sub> and δ<sup>13</sup>C-CO<sub>2</sub> data, and may be a pervasive feature of last glacial period CO<sub>2</sub> variability, but requires ground-truthing with additional, high-resolution MIS3 marine records.

304

## 305 **Conclusions**

Carbon isotope data from the last deglaciation and last glacial period clearly show that CO<sub>2</sub> variability is the sum of multiple mechanisms, many of which are triggered by abrupt climate change. During both periods millennial-scale variability is present and likely associated with the release of respired organic carbon from the deep ocean. Superimposed on these oscillations are two types of centennial-scale changes i) CO<sub>2</sub> increases and δ<sup>13</sup>C-CO<sub>2</sub> decreases in the middle of Heinrich stadials and ii) CO<sub>2</sub> increases and small changes in δ<sup>13</sup>C-CO<sub>2</sub> that are in-phase with rapid increases in NH temperature. During the deglaciation the millennial-scale component is enhanced and an additional carbon source is required to sustain the CO<sub>2</sub> rise through the entire deglaciation. This suggests that although abrupt climate variability is not the sole driver of the deglacial CO<sub>2</sub> rise, it may be a prerequisite. These potential links can now be tested with model experiments that use the MIS3 data to constrain the sensitivity to centennial and millennial-scale components and the deglacial data to evaluate how these mechanisms interact with changes in insolation, ice volume and global temperatures.

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## 319 **Acknowledgements**

This work was funded by NSF grants ANT 0838936 (Oregon State University), ANT 0839031 (Scripps Institution of Oceanography). Data will be made available online at National Climate Data Center: <https://www.ncdc.noaa.gov/paleo/study/24170>

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469 461.  
470

471 **Figure 1**

472 Taylor Glacier CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and  $\delta^{13}\text{C}$ -CO<sub>2</sub> (blue markers with black smoothing spline, *this study*).  
473 Panel A: NGRIP  $\delta^{18}\text{O}$ (North Greenland Ice Core Project Members, 2004). B: WDC  $\delta^{18}\text{O}$ (WAIS Divide  
474 Project Members, 2015). C: Hulu Cave  $\delta^{18}\text{O}$  (Cheng et al., 2016) D: WDC (Rhodes et al., 2015) (thin  
475 black line) and Taylor Glacier CH<sub>4</sub>. E: Siple Dome (Ahn & Brook, 2014)(gray markers) and Taylor  
476 Glacier CO<sub>2</sub>. F: Taylor Glacier  $\delta^{13}\text{C}$ -CO<sub>2</sub>. G: Talos Dome (Schilt et al., 2010) (gray markers) and Taylor  
477 Glacier N<sub>2</sub>O. H: Global Terrestrial O<sub>2</sub> isotopic fractionation,  $\Delta\epsilon_{\text{LAND}}$ , constrained from a combination of  
478 WDC and Siple Dome (Seltzer et al., 2017; Severinghaus et al., 2009). Dotted lines highlight the stadial  
479 to interstadial transitions and the red bar highlights the Heinrich Stadial with the associated rise in  
480 CO<sub>2</sub>, CH<sub>4</sub> and increase in  $\Delta\epsilon_{\text{LAND}}$ . The NGRIP chronology is increased by 0.63% prior to 22ka for  
481 consistency with the WDC chronology (WAIS Divide Project Members, 2015).

482

483 **Figure 2**

484 Cross-plot of atmospheric CO<sub>2</sub> and  $\delta^{13}\text{C}$ -CO<sub>2</sub> spanning the MIS3 section (blue, *this study*) and the last  
485 deglaciation, which is subdivide into the LGM to HS1 data (red) and late-Deglaciation (gray). Solid  
486 lines indicate linear regressions through the MIS3 (blue) and HS1 (red) data.

487

488 **Figure 3**

489  
490 Comparison of atmospheric record of  $\delta^{13}\text{C}$ -CO<sub>2</sub> with oceanic records of  $\delta^{13}\text{C}$  and nutrient utilization. A:  
491 Atmospheric  $\delta^{13}\text{C}$ -CO<sub>2</sub> (*this study*) on an inverted y-axis. B: The difference in  $\delta^{13}\text{C}$  between Sub-  
492 Antarctic mode water (SAMW) and circumpolar deepwater (CDW)(Ziegler et al., 2013). A stack of  
493 benthic  $\delta^{13}\text{C}$  (black line) from a combination of record in the deep Southern Ocean (grey circles)  
494 (Charles et al., 1996; Hodell et al., 2001, 2003; Ninnemann et al., 1999). Also indicated is the  
495 uncertainty on these chronologies (Barker & Diz 2014) based on synchronization between carbonate  
496 preservation and Greenland temperature (black boxes) and independent age control based on the  
497 Laschamp event (blue boxes). D: Foraminifera-bound  $\delta^{15}\text{N}$  indicating nitrate consumption in  
498 Subantarctic (Martínez-García et al., 2014). Red bars highlight the broad millennia-scale correlations.

499

500 **Figure 4**

501 Left Panel: Comparison of MIS3 data (*this study*) and Last Deglaciation (Bauska et al., 2016; Marcott et  
502 al., 2014; Rhodes et al., 2015) greenhouse gas variability. A: WDC CH<sub>4</sub> (Rhodes et al., 2015). B: Taylor  
503 Glacier N<sub>2</sub>O. C: Taylor Glacier CO<sub>2</sub>. D: Taylor Glacier  $\delta^{13}\text{C}$ -CO<sub>2</sub>. E: Pa/Th, a proxy for the strength of  
504 NADW formation (Henry et al., 2016; McManus et al., 2004)(orange), F: the opal flux in the  
505 SO(Anderson et al., 2009), a proxy for upwelling (green) (note the two different cores and axes are  
506 used in the comparison), G: the benthic to atmosphere <sup>14</sup>C age reconstructions for the Subantarctic  
507 zone(Gottschalk et al., 2016; Skinner et al., 2010), H: nssCa flux from the EDML ice core(Fischer et al.,  
508 2007), a proxy for dust delivery to Antarctica (brown), I: opal flux in the Subantarctic zone(Gottschalk



509 et al., 2016), a proxy for productivity (from the same core as the ventilation reconstruction). Yellow  
510 bars highlight the onset of interstadial conditions, black bars indicted the initial CO<sub>2</sub> rises and red bars  
511 correspond to the rapid jumps in CO<sub>2</sub> and CH<sub>4</sub>. Right Panel: Centennial-scale variability during the last  
512 glacial period from Taylor Glacier and WAIS Divide. Anomalies in the concentration and isotopic  
513 composition of the greenhouse gases are plotted with the relative timing determined by the mid-point  
514 in the CH<sub>4</sub> rise. Marker color corresponds to dashed lines in left panel.

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Figure 1.

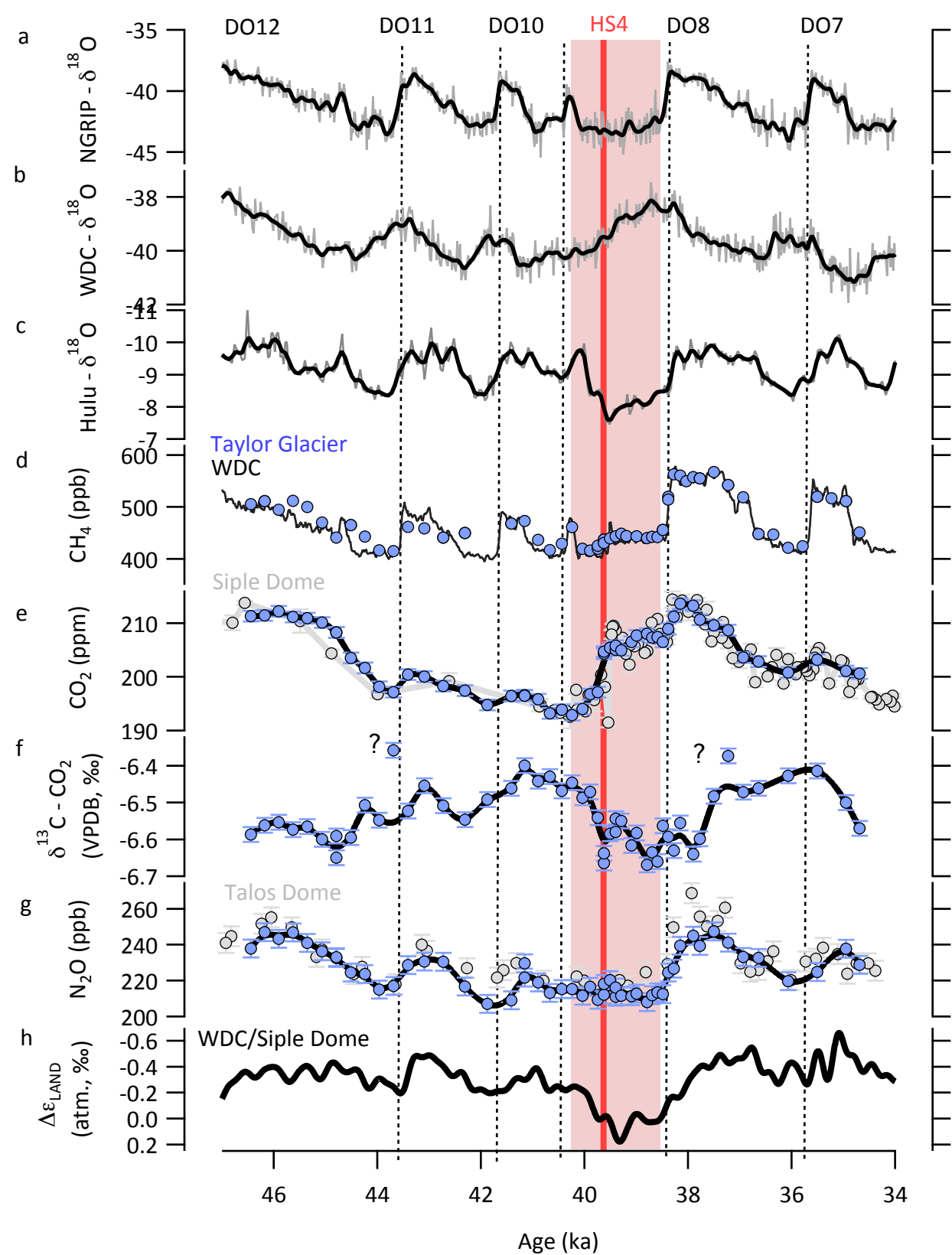


Figure 2.

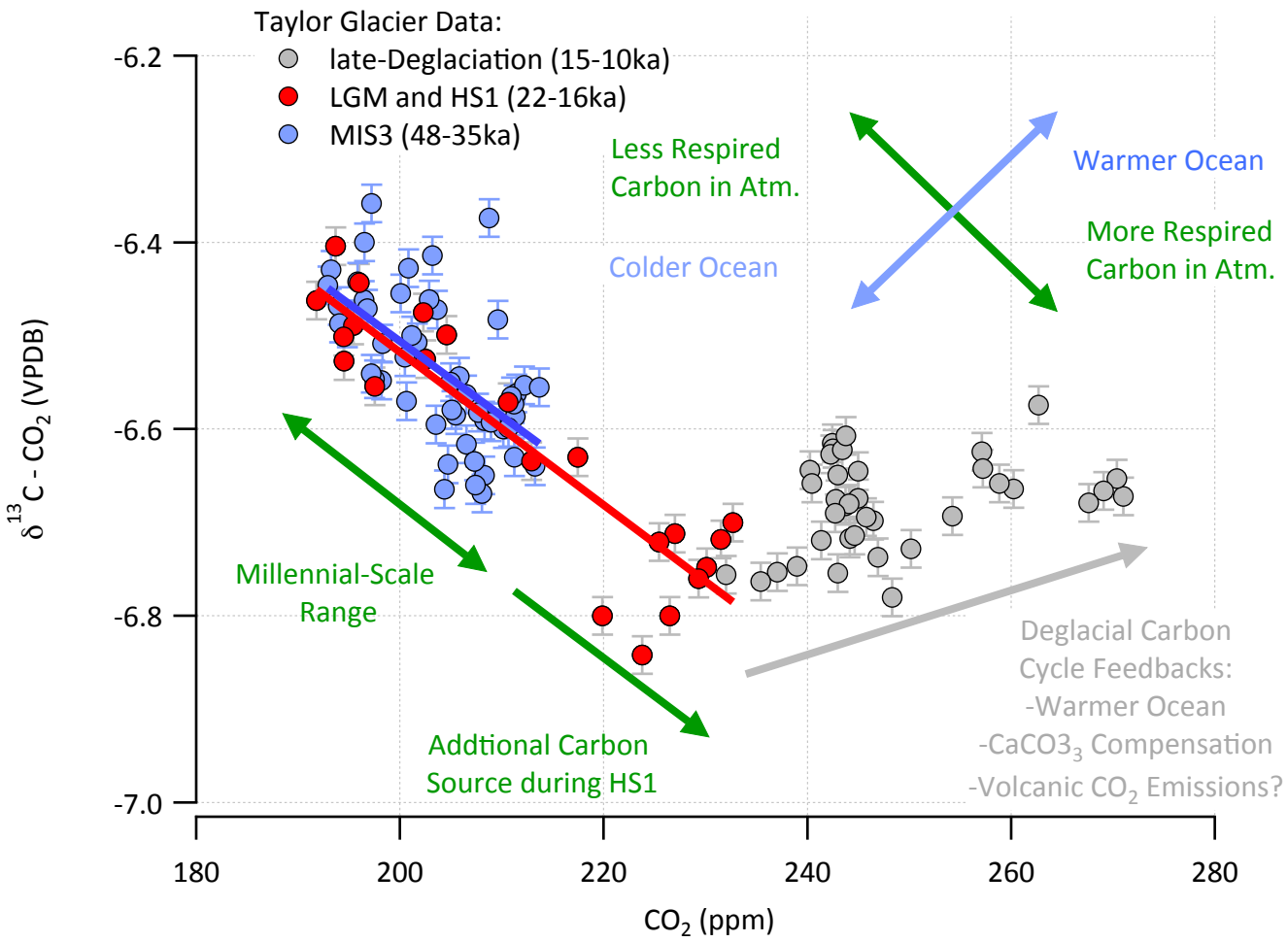


Figure 3.

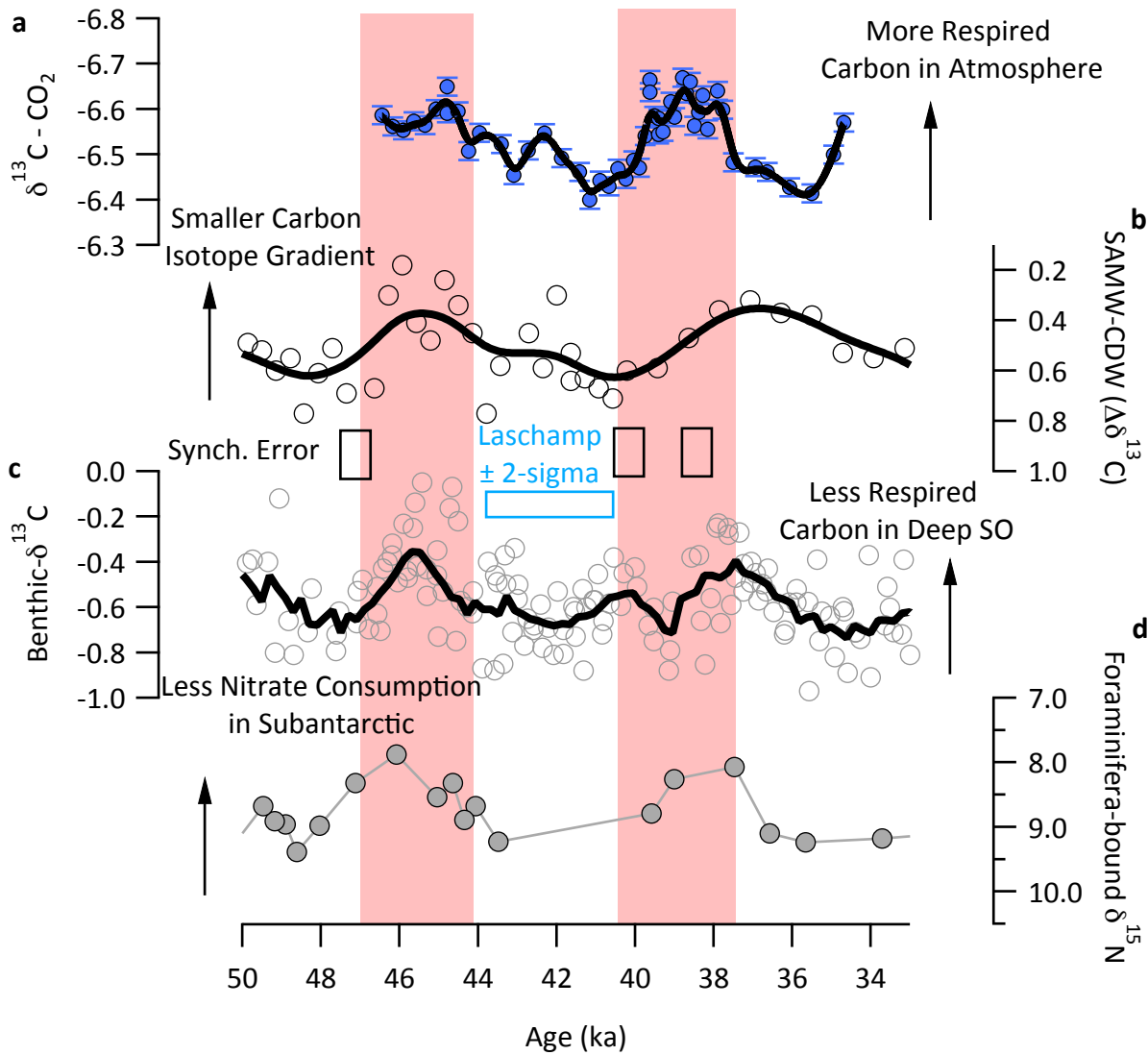


Figure 4.



